Supporting Information

Chromium Horseshoes as Polydentate Fluoride Ligands for Actinides and Group IV Metals

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Mass Spectrometry

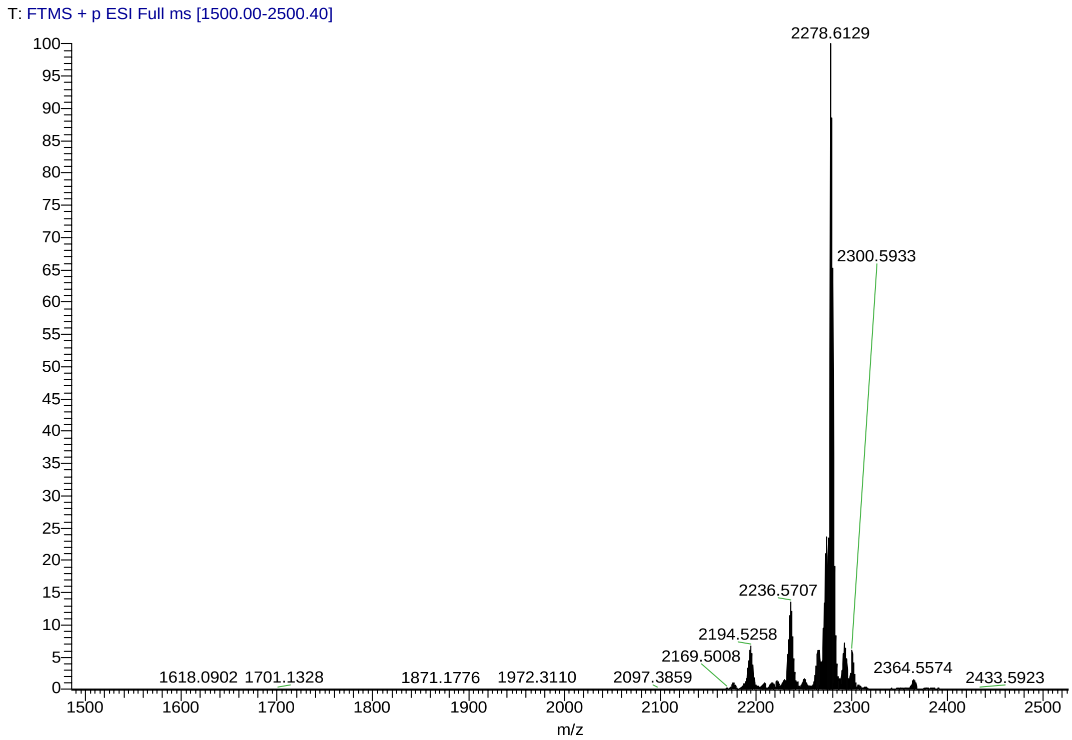

Figure S1. Electrospray mass spectrum of **2** (Cr_7Ti) showing the $[M+H]^+$ and $[M+Na]^+$ signals. ES-MS (THF, m/z): +2278 $[M+H]^+$; +2300 $[M+Na]^+$.

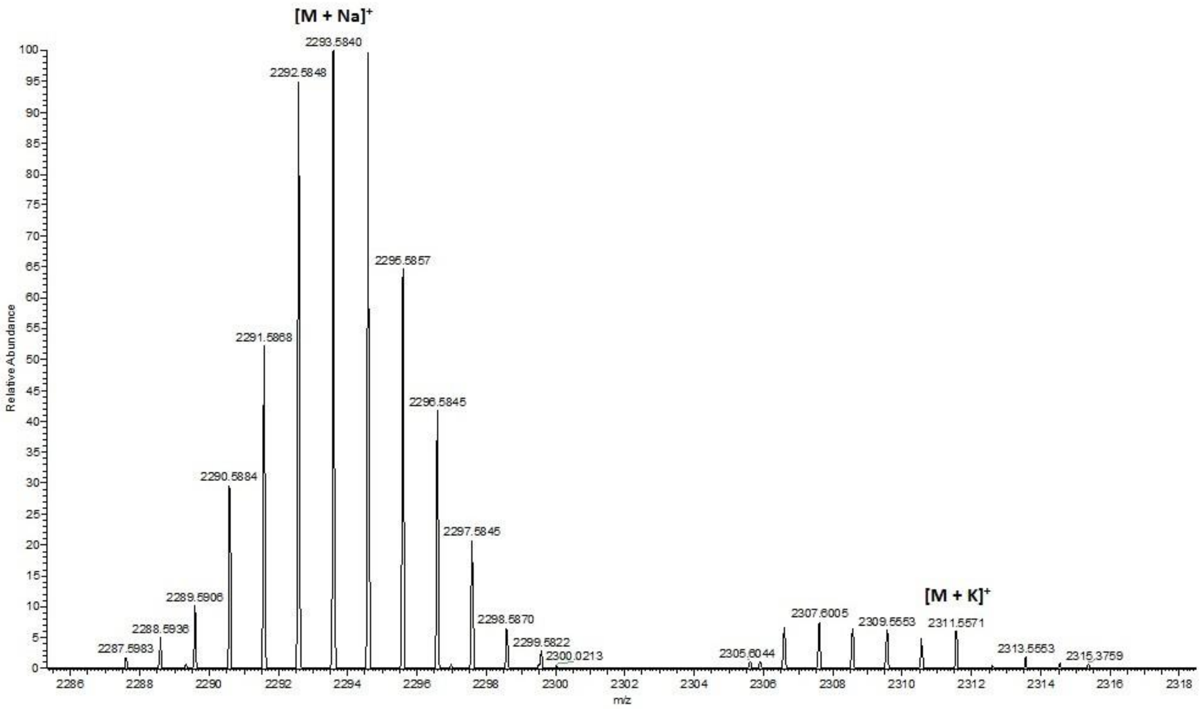

Figure S2. Electrospray mass spectrum of Cr_6Ti_2 showing the $[M+Na]^+$ and $[M+K]^+$ signals. ES-MS (THF, m/z): +2293 $[M+Na]^+$; +2311 $[M+K]^+$.

Additional magnetic measurements

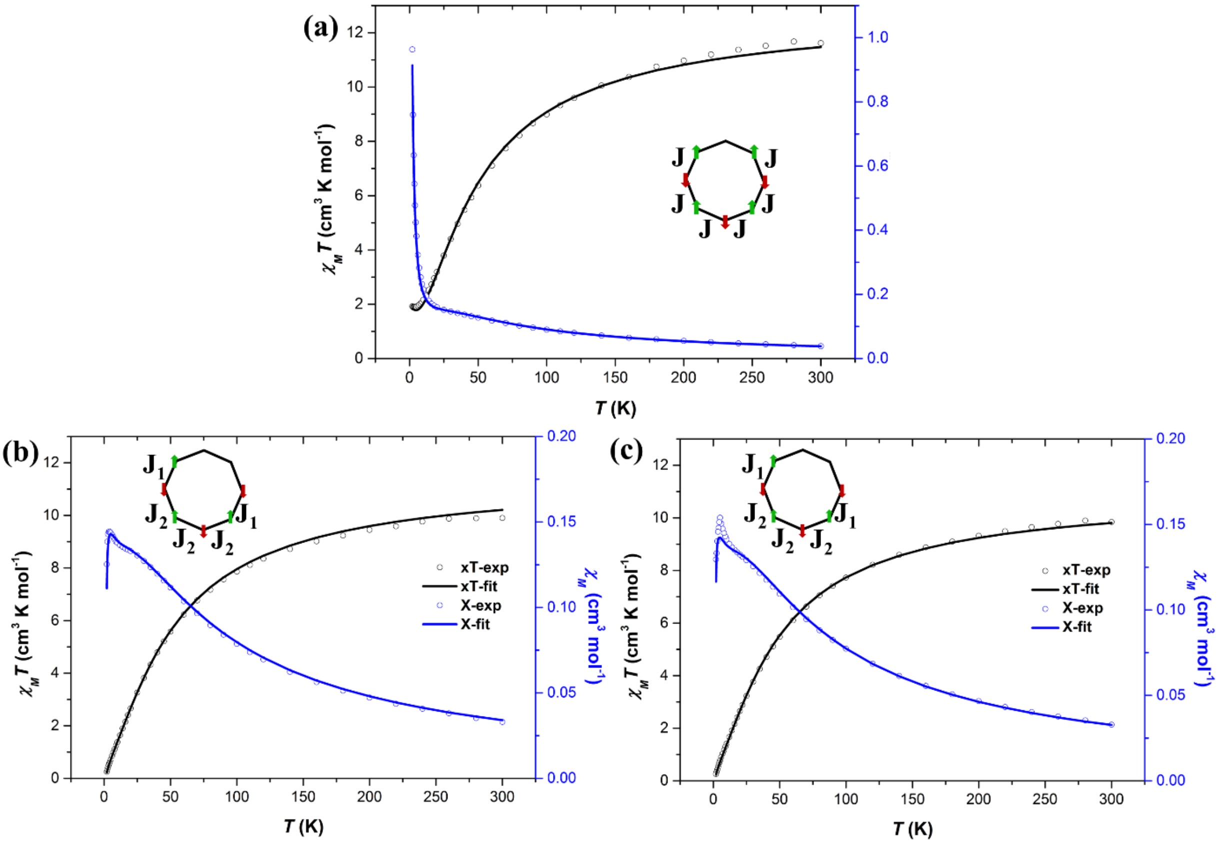

Figure S3. Susceptibility vs. Temperature of (a) **2**, (b) **3** and (c) **5**. $\chi_M T$ *vs. T*, black circles and χ_M vs *T*, blue circles. Solid lines represent fittings, where g_{cr} = 1.98, (a) **2**, J = -5.7 cm^{-1} (b) **3**, J_1 = -5.8cm^{-1} and J_2 = -5.3 cm^{-1} and (b) **5**, J_1 = -5.3 cm^{-1} and J_2 = -6.2 cm^{-1}. Magnetic coupling models are presented as insets.

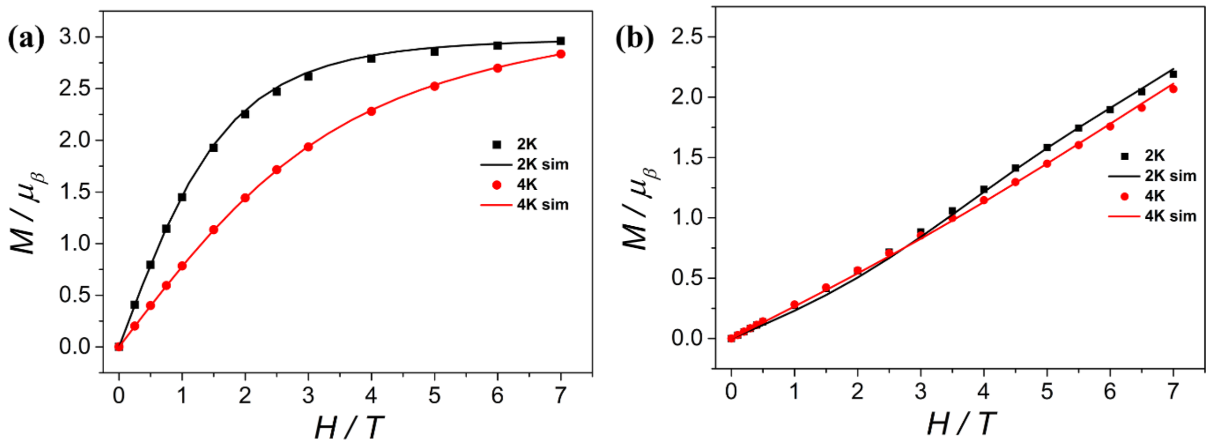

Figure S4. Field-dependent Experimental Magnetization plots at indicated temperatures for (a) **2** and (b) **5**. Solid lines represent simulations using the parameters derived from fitting the susceptibility data.

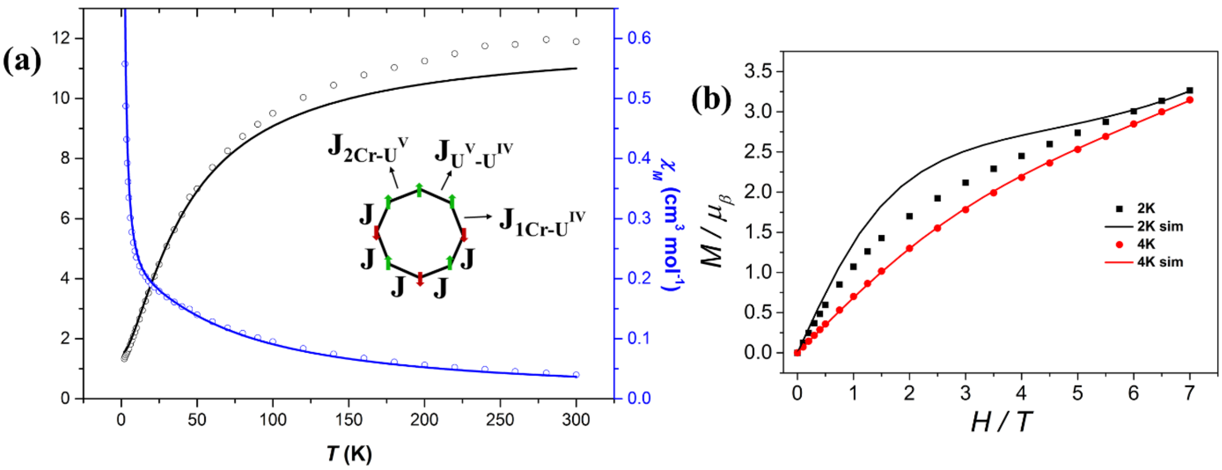

Figure S5. Susceptibility *vs.* Temperature and Field-dependent Experimental Magnetization plots of **6**. Solid lines represent fittings, where g_{cr} = 1.98, g_U^{IV}=1.70 and g_U^{V}=2.00, J=J_{Cr-Cr} = -5.2 cm^{-1}, J_{1Cr-U}^{IV} = -2.1 cm^{-1}, J_{2Cr-U}^{V} = +4.5 cm^{-1}and $J_{U^V-U^{IV}}$ = +0.8 cm^{-1}.

Additional EPR Spectra

Spectral simulations were performed using the EasySpin 5.0.2 simulation software.

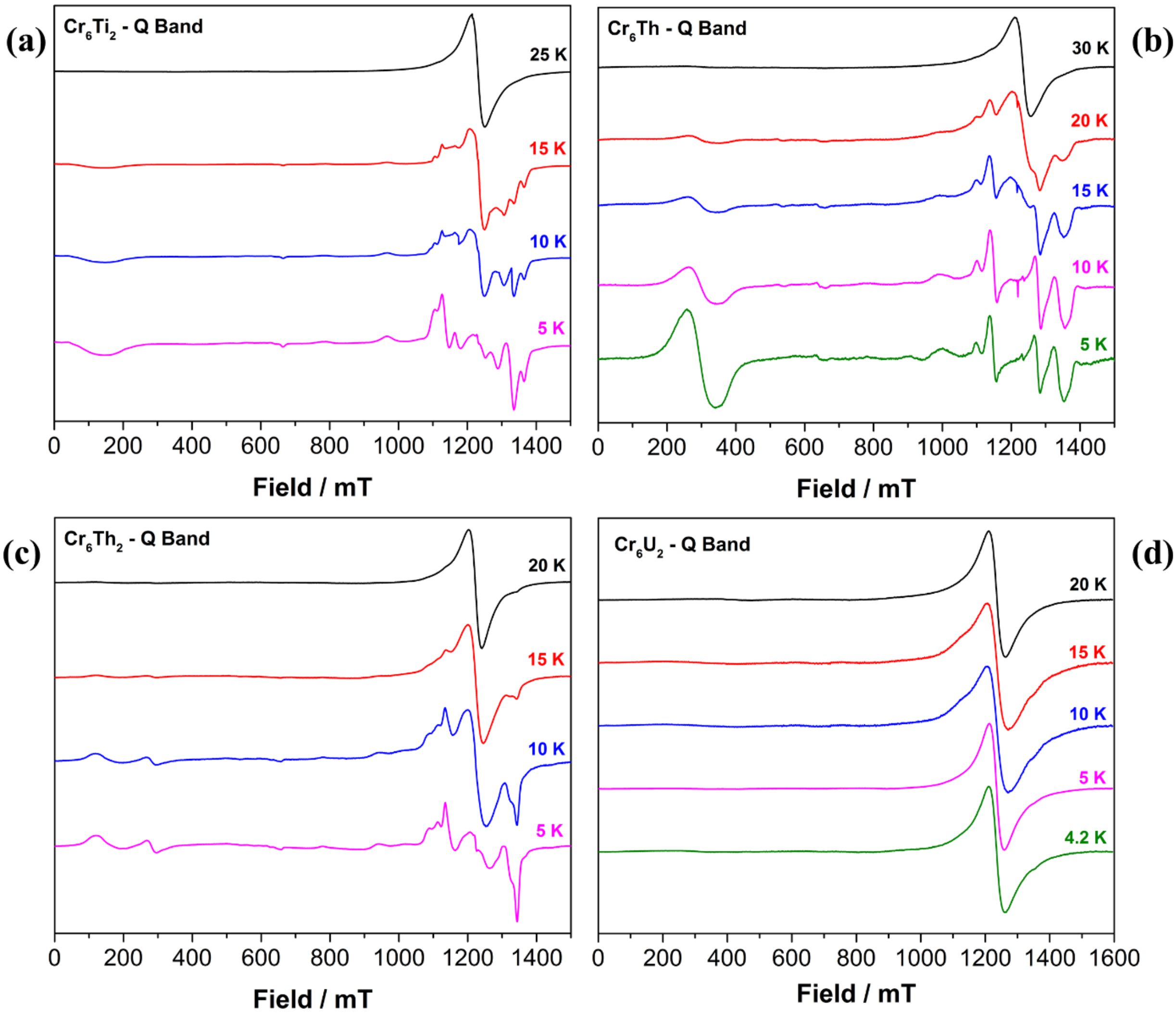

Figure S6. Variable temperature experimental Q-band (ca. 34 GHz) EPR spectra for grounded powder samples of (a) **3** at 33.9705 GHz, (b) **4** at 34.1771 GHz, (c) **5** at 34.1434 GHz and (d) **6** at 34.1266 GHz.

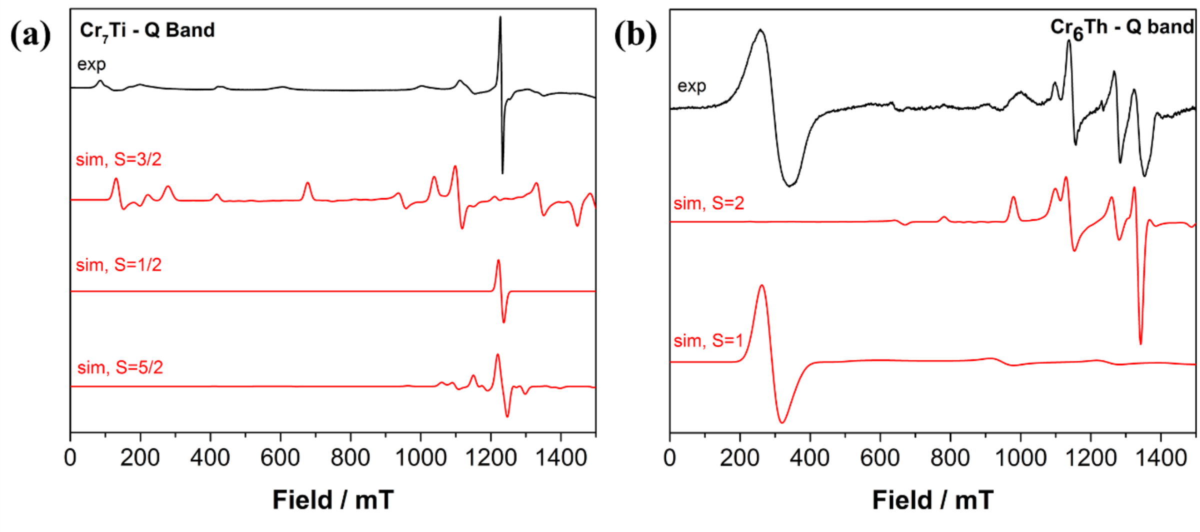

Figure S7. Experimental Q-band (ca. 34 GHz) EPR spectra (black) and simulations (red) for (a) **2** and (b) **4** and simulations of the individual spin states used for the sum simulation presented in the main text.